

# STUDY OF GEOMETRIC EFFECT TO THE CREVICE CORROSION BEHAVIOUR IN STAINLESS STEEL

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## ABSTRACT

This thesis deals with the geometric effect assessment to the crevice corrosion behaviour in stainless steel with a variations of crevice formers of different gap and depth dimensions. The objective of this thesis is to investigate the crevice corrosion behaviour of AISI 304 stainless steel in a variation of crevices geometry at room temperature. The thesis describes the effect of the geometric dimension on the crevice mouth to the AISI 304 stainless steel. This thesis also aimed to identify the crevice corrosion behaviour and to predict the fatigue life and identify the critical locations of the components. AISI 304 stainless steel were studied in this thesis which commonly used in industry. The crevice formers with variation dimension of gap and depth was machined in the laboratory. The method to analyse the corrosion rate and corrosion damage for the AISI 304 stainless steel is by weight loss. The observed weight loss data is then analysed by using corrosion rate equation accordance to American Society for Testing Material G1. From the results, it is observed that the amount of weight loss due to crevice corrosion behaviour increased as the crevice mouth opening increase where else at smaller crevice mouth opening. The result also can significantly shows the result of the differences between the general corrosion and the crevice corrosion; and also the corrosion behaviour inside the different in crevice gap and depth dimension.

## ABSTRAK

Tesis ini membentangkan penilaian kesan geometri untuk kelakuan kakisan ceruk dalam keluli tahan karat dengan variasi pembentuk celah jurang yang berbeza dan dimensi mendalam. Objektif projek ini adalah untuk menyiasat tingkah laku celah kakisan AISI 304 keluli tahan karat dalam perubahan celah geometri pada suhu bilik. Tesis ini menerangkan kesan dimensi geometri pada mulut ceruk kepada AISI 304 keluli tahan karat. Tesis ini juga bertujuan untuk mengenal pasti tingkah laku hakisan ceruk dan untuk meramal hayat lesu dan mengenal pasti lokasi-lokasi kritikal komponen. AISI 304 keluli tahan karat telah dikaji dalam tesis ini yang biasa digunakan dalam industri. Pembentuk celah dengan dimensi perubahan jurang dan mendalam telah dimesin di makmal. Kaedah untuk menganalisis kadar hakisan dan kerosakan hakisan untuk AISI 304 keluli tahan karat adalah dengan penurunan berat badan. Data berat badan diperhatikan kemudiannya dianalisis dengan menggunakan kadar kakisan persamaan menurut Persatuan Amerika untuk Ujian G1 Bahan. Daripada hasil kajian, didapati bahawa jumlah kehilangan berat badan kerana celah kelakuan kakisan meningkat peningkatan membuka mulut ceruk mana lagi di celah kecil membuka mulut. Hasilnya juga ketara boleh menunjukkan hasil daripada perbezaan antara karat umum dan hakisan ceruk dan juga kelakuan kakisan di dalam jurang yang berbeza dalam celah dan dimensi mendalam.

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**LIST OF SYMBOLS**

$b_a$	Tafel slope of anodic polarization curve (mV/decade)
$b_c$	Tafel slope of cathodic polarization curve (mV/decade)
$E$	Electrode potential (mV)
$E_{A/P}$	Active / passive potential (mV)
$E_{surf} \text{ or } E_{app}$	Applied potential (mV)
$E_{rev}$	Reversible Potential (mV)
$E_{corr}$	Corrosion Potential (mV)
$F$	Faraday's Constant (96485 C/mol)
$M_w$	Molecular weight (g/mol)
$I$	Current ( $\mu\text{A}$ )
$i_{corr}$	Anodic current density ( $\mu\text{A}/\text{cm}^2$ )
$L$	Crevice depth (cm)
$R_p$	Polarization resistance ( $\Omega \text{ cm}^2$ )

## LIST OF ABBREVIATIONS

AISI	American Institute of Steel and Iron
ASTM	American Standard of Testing and Material
D	Density
FYP	Final year project
ipm	Inches per month
ipy	Inches per year
LPR	Linear polarization resistance
MGC	Multiplexed galvanic corrosion
ml	Milliliter
mm	Millimeter
mpy	Mile per year
MPP	Multiplexed potentiodynamic polarization
OCP	Open circuit potential (mV)
SEM	Scanning electron microscope
SS	Stainless steel (AISI 304)
T	Temperature (°C)
t	Time (h)
UMP	Universiti Malaysia Pahang
ZRA	Zero Resistance Ammeter

## **CHAPTER 1**

### **INTRODUCTION**

#### **1.1 INTRODUCTION**

This chapter explains about the background of study, problem statement objectives, and scopes of this study. The purposes of this study can be identified by referring at the problem statement of this study. Besides that, the detail of the study and output can be obtained based on the objectives and scopes of this study.

#### **1.1 BACKGROUND OF STUDY**

Stainless steel is one of the world's most used metals with a huge consumption every year. Its strength, conductivity, barrier properties and its excellent corrosion resistance is the most important advantages and the main reasons for the continued growth in the usage of stainless steel. Stainless steel and its alloys are widely used in industrial systems due to their excellent corrosion resistance properties. However, they are more susceptible to localized corrosion than uniform corrosion in aggressive environments (Kabi, 1985). Crevice corrosion is a type localized corrosion that can happen in some good anti-corrosion materials such as stainless steel and it is depend on the presence of a

protective layer of oxide on the stainless steel surface. It is possible for this oxide layer to break down under certain conditions such as in reducing acids where the atmosphere is reducing (Stainless Steel for Design Engineer, 2008).

The way of the components are designed for example under gasket, overlapping surfaces or in associated with incomplete weld penetration sometimes resulting the areas of which the oxide layer. Apart from the passive films characteristic, there are several factors that are responsible for crevice corrosion initiation in stainless steel such as alloy composition, mass transport in and out of crevices and the geometrical aspects. All of these conditions can promote to all form crevices to initiate corrosion. It is very important to properly design the component especially on the geometrical aspect to avoid crevice corrosion to happen because crevice corrosion may lead to sudden devastating failure of the metal in service. Hence, in order to study the crevice corrosion behaviour with respect to the geometrical aspect, the development of crevice former with different gap and depth dimension is important to study more detail on the crevice corrosion behaviour.

## **1.2 PROBLEM STATEMENT**

Crevice corrosion is a form of localized attack that occurs frequently on metals exposed to stagnant solutions within shielded areas such as gaskets, lap joints, holes and crevices under bolts. This form of corrosion is usually very difficult to detect, predict and design against due to the size and locations of the corroding crevice. Materials with high corrosion resistance are usually the most vulnerable to this form of corrosion. Well known examples of susceptible metals are stainless steel alloys. Crevice corrosion leads to the breakdown of the passive film in these localized areas on stainless steels is believed to result from number of factors that are environmental, metallurgical, electrochemical and geometrical in nature. Geometrical aspects such as the exterior to interior crevice area ratio, width and depth of the crevice are also known to contribute to this corrosion. Thus, it is essential this study to investigate the effect of specimen geometry to the crevice corrosion behaviour to minimizes its effect that the metal in service.

### **1.3 OBJECTIVES**

The main objective of this study is to investigate the crevice corrosion behaviour of AISI 304 stainless steel in a variation of crevices geometry at room temperature.

### **1.4 SCOPES**

The scopes for this project are as follows:

- i. The material used is stainless steel (AISI 304).
- ii. Variation of gap (1 mm, 2 mm, 4 mm and 6 mm) and depth (0.5 mm, 1.0 mm and 2.0 mm) of the crevices are used as manipulated parameters.
- iii. Microstructure analysis of the different specimen geometry by using Scanning Electron Microscope (SEM).
- iv. Corrosion rate is assessed by weight loss method.
- v. Corrosion test of AISI 304 stainless steel is done in 3.5wt% NaCl and 50% CH<sub>3</sub>COOH to imitate oil and gas pipeline environment.

## **1.6 THESIS OUTLINE**

This thesis consists of 5 chapters which illustrate the flow of the project from introduction to the conclusion. Every chapter has its own specific contents. The viewer may understand the detail of this project after they read this thesis and obtains the output of this project.

Chapter 1 comprised of the overview of the chapter, the background of study, problem statement, objectives and scopes of this study.

Chapter 2 contained all the literature review and some presentation of the earlier work.

This chapter also will further explained the brief explanation of the material used, corrosion related information from others study that is related and relevant to this study.

In the Chapter 3 contained research methodology of this project. This chapter also explained the about the flow work of the study and also on how the project is conducted.

Chapter 4 contained the result and discussion than can be obtained by the end of the study. This chapter also explained the analysis and discussion of the result. The result obtained is discussed and compared to previous study.

Chapter 5 concluded about the project. This chapter also includes the future recommendation of the project.

## **CHAPTER 2**

### **LITERATURE REVIEW**

#### **2.1 INTRODUCTION**

This chapter presents a brief overview of the stainless steel used and their application in industrial systems. Furthermore, the theories that initiate crevice corrosion, experimental techniques used for previous studies and also the experimental techniques to be used in this work are explained in detail.

#### **2.2 STAINLESS STEEL**

One of the reason stainless steels are used because of its corrosion resistance properties even though they do suffer from certain type of corrosion in some environment. Hence, care must be taken to select a suitable stainless steel grade according to the application this is because corrosion that could happen in stainless steel can cause variety problems depending on their applications (Kadry, 2008). The variety problems stated, can be in the form of the failure of the structure build due to unsuitable grade of stainless steel used in specific industry. This kind of problem must be avoided in order to reduce the huge cost involved in building equipment for industrial systems purposes. Because of their unusual resistance to corrosion due to high chromium content oxide film on the surface, this high temperature steels applied in many industry. The passive oxide film

form is usually very thin and protective in oxidizing environments. These protective environments can be air and water (Etor, 2009). The application of stainless steel can be very wide and this is shown in Table 2.1.

Stainless steel can be classified into several categories such as martensitic, ferritic, austenitic and duplex groups (Sedriks, 1996). However, the purposes of this study, the emphasis will be placed on the AISI 304 stainless steel which is an austenitic stainless steel. Austenitic stainless steel contained about 18% of chromium and 8% of nickel in which contribute to their high resistance of corrosion. The most common grade is grade AISI 304 stainless steel for food and surgical stainless steel uses, chemical processing, the pulp and paper industry, and also part that is exposed to marine atmosphere and tubing. Due to its affordability and its ability to form protective oxide film on its surface for corrosion resistance in oxidizing environments, AISI 304 stainless steel is widely used in the past and it is commercial grade of austenitic stainless steel.

### **2.2.1 Austenitic Stainless Steel**

Austenitic stainless steels are well known for their good mechanical properties and corrosion resistance characteristics. It is also having an austenitic, face – centered cubic (fcc) crystal structure. They are ductile, tough, and easy to form and weld. These alloys are mostly common austenitic alloys are Fe-Cr-Ni steels as 300 series. They also offer corrosion resistance with sufficient amount of chromium and with nickel that used to maintain the austenitic form of the stainless steel at room temperature (Shirazi, 1994). AISI 304 austenitic stainless steel composition and, mechanical properties is shown in Table 2.2 while Table 2.3 is showing the chemical composition and the mechanical properties of some austenitic stainless steel.



**Table 2.1:** Application of stainless steels under variety of environments

<b>Field/Industry</b>	<b>Applications</b>
<b>Consumer goods</b>	Kitchenware and tableware, kitchen sinks, laundry equipment and electrical and electronics appliances.
<b>Architecture, building &amp; construction</b>	Roofing and guttering, signage, curtain wall supports, bridges, barrier walls and decking.
<b>Food processing</b>	Beer and wine fermenters, fruit juice tanks and piping storage vessels.
<b>Transportation</b>	Exhaust system of the automotive, seagoing chemical tankers and passenger railcars.
<b>Chemical</b>	Heat exchangers, high temperature furnace equipment, and components for nuclear reactors.

Source: Etor (2009)

**Table 2.2:** Chemical composition and mechanical properties of AISI 304 austenitic stainless steel

%C	%Cr	%Ni	%Mo	Tensile Strength (MPa)	0.2% Yield Strength (MPa)	Elongation % in 50.8 mm	Hardness (HB)
<b>0.08</b>	18-20	8-10	-	579	290	50	201

Source: AKSteel (2007)

**Table 2.3:** Chemical composition and mechanical properties of some austenitic stainless steel

AISI type	%C	%Cr	%Ni	%Mo	Tensile Strength (MPa)	0.2% Yield Strength (MPa)	Elongation % in 50.88mm	Hardness (HB)
<b>304</b>	<b>0.08</b>	<b>18-20</b>	<b>8-10</b>	<b>-</b>	<b>205</b>	<b>515</b>	<b>40</b>	<b>201</b>
<b>316</b>	0.08	16-18	10-14	2-3	205	515	40	217
<b>316L</b>	0.03	16-18	10-14	2-3	170	485	40	217

Source: Shirazi (1996)

With excellent corrosion resistance characteristic plus its good mechanical properties at elevated temperatures, these alloys also widely used in nuclear reactors applications (Weiss and Stickler, 1972). Material Type AISI 304 stainless steels exhibit excellent corrosion resistance as they are providing excellent corrosion resistance to most chemicals environment, textile production industries and petroleum industries. Grade AISI 304 stainless steel is generally withstands ordinary corrosion in architecture, it is durable in typical food processing environments and resists some chemical. The crevice

corrosion behaviour has been studied by various researchers (Malik and Fozan, 1993; Abdulsalam and Pickering, 1998; Naganuma et al. 2010; Matsushima et al. 1978).

### **2.2.2 Crevice Corrosion of Austenitic Stainless Steel.**

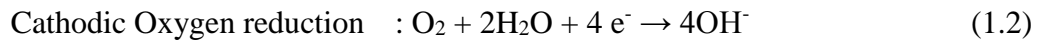
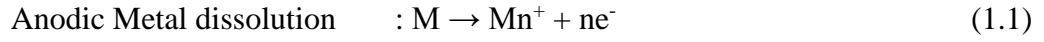
Crevice corrosion commonly occurs in austenitic stainless steel that develops a barrier or passivation layer. There are three conditions for this passivation layer to develop. The external metal outside the crevice and the metal must be electrically connected is the first condition. The second condition, there is stagnant solution and provide an ionic path to the solution near the metal outside the crevice. The third condition is the external metal surface must be passive (Etor, 2009).

All shapes and joints which may form crevices in the stainless steel design equipment, should be avoided. Area within crevice, the oxygen does not have ready access to and these may becoming anodic to the remaining exposed metal outside, the extent of the attack within crevice has been shown to be proportional. By completely sealing such crevices, or altering design eliminated, this form of corrosion of stainless steel may be eliminated or reduced to minimum. For example, if gaskets are used, they should be pulled up tightly over and non-porous over their entire area (Inco, 1963).

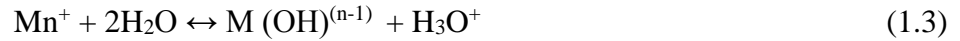
## **2.3 MECHANISM OF CREVICE CORROSION**

Various researchers have made attempts to examine the problems of crevice corrosion as well as propose mechanisms for its progression (Fontana and Greene, 1967; Kain, 1979; Oldfield and Sutton, 1978; Lee, 1981; Alavi and Cottis, 1985; Pickering, 1993; Wolfe, 2006; Bocher *et al.* 2008). The reproducibility of the results, namely, the corrosion initiation and propagation rate, has always been a problem in crevice corrosion studies. The crevice corrosion mechanism is complex due to its dependence upon many factors, and is included in the discussion of crevice corrosion initiation and propagation stages.

Given a metallic crevice in an oxygen - saturated electrolytic solution at neutral pH, metal dissolution and the accompanying cathodic oxygen reduction to hydroxide ions occurs uniformly over the metal surface including in the interior of the crevice. These reactions are represented as in equation (1.1) and (1.2) during the initiation stage (Fontana and Greene, 1967).



Eventually, the oxygen within the crevice gets depleted due to mass transfer limitations created by the crevice geometry, while the oxygen in the bulk solution does not deplete. This creates a differential aeration cell where the bold area external to the crevice acts as the cathodic site and the crevice area, with less or no oxygen, acts as the anodic site with metal dissolution. Furthermore, metal ions are transported slowly out of the crevice by diffusion or migration, and hydrolysis of these ions leads to a gradual pH decrease in the crevice as represented in equation (1.3).



The conduction of these charged metal ions through the electrolyte may cause a significant drop in potential, commonly referred to as the *IR* drop (Pickering *et al.* 1993). Electro-neutrality requires the migration of anions such as  $\text{OH}^-$  and  $\text{Cl}^-$  ions towards the crevice, but the  $\text{OH}^-$  precipitates as a metallic hydroxide,  $\text{M}(\text{OH})_n$  before reaching the crevice.



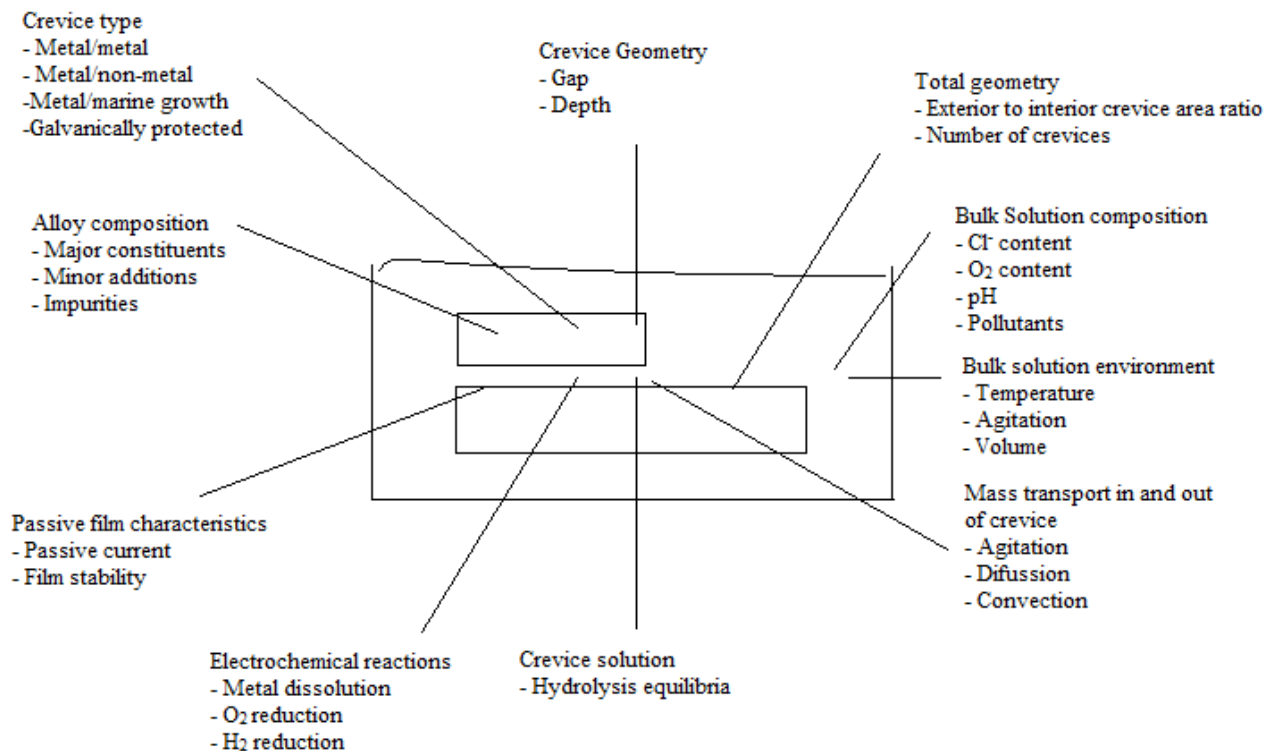
The  $\text{Cl}^-$  ions are attracted to the anodic metal surface, and this lead to the development of an aggressive acid chloride local solution within the crevice. At the end of the incubation phase of crevice corrosion, a high concentration of hydrogen ions have accumulated in the crevice due to hydrolysis of dissolution products, and there is a build-up of chloride ions in the crevice. This may lead to the loss of stability of the passive film

protecting the metal. This facilitates anodic dissolution and leads to a more rapid breakdown of these films on the metal.

However, the external area of the metal sample, where cathodic reactions occur, still remains passive during this process (Laycock *et al.* 1997; Betts and Boulton, 1993). As dissolved species build up within the crevice, high anodic current is observed towards the mouth of the crevice as supported by the bold surface cathodic reactions. This increase in anodic current close to the crevice mouth is attributed to the lower IR drop between the crevice mouth and the bold surface. As the distance from the crevice mouth increases, larger IR drop is established between the crevice mouth and the crevice tip, and if the potential drop at the crevice tip becomes too large, the net anodic current produced at locations deeper into the crevice will cease due to mass transfer limitations (Betts and Boulton, 1993; Kennell *et al.* 2009). Finally, the accumulation of corrosion products at the crevice mouth over time may cause a significant decrease in the corrosion rate by introducing large resistance between the crevice and the external bold surface.

## **2.4 GEOMETRIC EFFECT AS A FACTOR AFFECTING CREVICE CORROSION**

There are several factors that are responsible for crevice corrosion initiation in stainless steel. Such factors can be environmental in nature and they include bulk and crevice solution oxygen content, pH, chloride level and temperature. Passive films characteristics, alloy composition, mass transport in and out of crevices also can influence crevice corrosion to occur. Geometrical aspects such as the exterior to interior crevice area ratio, width and depth of the crevice are also known to contribute to this corrosion. The initiation of crevice corrosion is generally controlled by the passive current and the crevice area ratio is likely to be high in practical situation (Dayal, 2002).



**Figure 2.1:** Various factors influencing crevice corrosion

Source: Dayal (2002)

Oldfield and Sutton (1978) showed that crevice corrosion initiation time in seawater increased as the crevice gap dimension increased from  $0.1\mu m$  to  $1\mu m$ . A similar study proposed by Kain (1983 and 1985) that for a crevice depth of  $0.1cm$ , a gap less than  $0.01\mu m$  is might be required for the crevice initiation of AISI 304 SS in natural seawater.

Dayal et al. (1983) showed that crevice corrosion initiation time is strongly dependent on the depth to width ratio of the crevice geometry. By increasing the crevice gap width, this will increase the initiation time of the corrosion to occur. According to Alavi and Cottis (1987), a wider crevice gap reduces the chloride ion activity in the crevice and hence this will increase its initiate time for the corrosion to happen. Based on the previous studies, due to oxygen diffusion restrictions, the tighter of the crevices would be necessary for crevice corrosion to initiate at lower chloride.

Kennell et al. in 2009 used critical crevice corrosion scaling laws to model crevices dimensions that will become active upon oxygen depletion, become active after an incubation period or not become active at all. The crevice lengths modelled were between 1 cm and 8 cm, and the crevice widths between 1  $\mu\text{m}$  to 1 mm. The corrosion processes were modelled for extended period of time for specific length and width during each simulation. From the model, immediate crevice corrosion was observed for crevice length above 1 cm with crevice width of 100  $\mu\text{m}$ . For the crevice length above 1 cm and crevice width  $> 100 \mu\text{m}$ , the crevice became active after a period of incubation, and crevice length below 1 cm for all crevice width did not become active.

The critical aspect ratio,  $AR_c$ , was observed at the boundary between the active crevice zone and not-active  $v = \text{crevice zone}$ , where the crevice scaling factor,  $L^2/G = 401 \text{ cm}$ . However, in contrast to expectation, Pickering (2003) observed that crevice corrosion will occur faster in crevice of large opening or gap dimension,  $a = 0.05 \text{ cm}$  and applied potential  $> 350 \text{ mV}$ . It was predicted that the wider the gap will creating less IR drop in the crevice electrolyte than a tighter gap, thus more chances of crevice corrosion initiation. A similar view studied by Abdulsalem (2007). He observed that for a crevice gap of 0.03 cm, an application of  $E_{surf} = 530 \text{ mV}$  on nickel in various sulphuric acid solutions caused the initiation crevice corrosion at varying depth inside the crevice. Pickering et al. (1998) work showed 0.04 cm as the upper limit crevice width for crevice corrosion initiation in a 1 N Sulphuric acid solution.

Alkire and Lott (1985) reported that no crevice corrosion was observed for type 304 stainless steel in 0.1 N NaCl solutions with large crevice gaps of  $> 7.5 \mu\text{m}$ , low manganese sulphide inclusion densities and an applied potential of  $< 75 \text{ mV}$ . However, with tighter crevice of  $7.5 \mu\text{m}$ , higher inclusion densities and applied potentials of  $= 75 \text{ mV}$ , crevice corrosion was observed to occur. They therefore inferred that crevice corrosion did initiate due to pit formation through dissolution of the manganese sulphide inclusions.

Eun-Young Na et al. (2005) investigated the initiation of crevice corrosion for ferritic stainless steel by measuring the potentials and current change in the crevice using

a depth profile technique with a micro capillary tube. The measured potentials at various crevice widths were lower than the external surface potential of the metal due to the IR drop between the opening and bottom of the crevice. The crevice corrosion developing time for 0.1 mm and 0.2 mm widths were 15 minutes and 23 minutes respectively.

However, the larger crevice width of 0.5 mm showed no evidence of crevice corrosion initiation. This supports the view that crevice corrosion will not initiate in crevice widths larger than 0.4 mm (Pickering et al. 1998).

## 2.5 ARTIFICIAL CREVICE STUDIES

Single and multiple crevices have been studied by various researchers such as Hu *et al.* (2011), Klassen et al, (2011), Al-ameer and Uthman (2011) and Abdulsalem (2005)

### 2.5.1 Crevice corrosion in 13Cr stainless steel

Hu *et al.* (2011) studied the crevice corrosion behaviour of 13Cr stainless steel in NaCl solution. They investigated mainly by using electrochemical noise measurements. The crevice opening dimension ( $a$ ) and the area ratio of the electrode outside the crevice to the inside the crevice ( $r$ ) is considered. The crevice used in their experiment was built from a 13Cr stainless steel with exposure area of 1.0 cm<sup>2</sup>, 19 cm<sup>2</sup>, or 38 cm<sup>2</sup> as a working electrode was machined into rod and sealed with epoxy resin as shown in Figure 2.2.

The exposed surface of working electrode (WE) was ground to 1200 grit with silicon carbide paper and then cleaned with distilled water and degreased with acetone. The 3.5 wt.% of NaCl solution (pH 6.7) were used. Two working electrode were used. One with an exposure area of 1.0 cm<sup>2</sup> was placed in crevice solution as Working Electrode 1, the other working electrode with exposure area of 1.0 cm<sup>2</sup>, 19 cm<sup>2</sup>, or 38 cm<sup>2</sup> was immersed in the bulk solution and which resulting in various  $r$  values.